

[Document Name] CLAIMS

[Claim 1]

A process for production of a purified green tea extract containing from 25 to 90 wt% of non-polymer catechins based on a dry weight of said extract, which comprises dispersing a green tea extract in a 91/9 to 97/3 by weight mixture of an organic solvent and water, and bringing the resulting dispersion into contact with activated carbon and acid clay or activated clay.

[Claim 2]

The process according to claim 1, wherein the organic solvent is ethanol.

[Claim 3]

The process according to claim 1 or 2, wherein the weight ratio of the non-polymer catechins to caffeine in the purified green tea extract is from 25 to 200.

[Claim 4]

A purified green tea extract, wherein the purified green tea extract comprises from 40 to 90 wt% of non-polymer catechins based on a solid content of said purified green tea extract, and the weight ratio of the non-polymer catechins to caffeine is from 25 to 200.

[Document Name] SPECIFICATION

[Title of the Invention] PRODUCTION PROCESS OF PURIFIED GREEN  
TEA EXTRACT

[Field of the Invention]

[0001]

This invention relates to a process for producing, from a green tea extract, a purified green tea extract containing non-polymer catechins at high concentration.

[Background of the Invention]

[0002]

Catechins are known to have a suppressing effect on increase in cholesterol level and an inhibitory effect on  $\alpha$ -amylase activity (see, for example, Patent Document 1 and Patent Document 2). To develop such physiological effects of catechins, it is necessary for an adult to drink 4 to 5 cups of tea per day. Accordingly, there is a desire for a technology that permits addition of catechins at a high concentration to beverages to facilitate ingestion of a large amount of catechins.

[0003]

In tea leaves, however, caffeine component is also contained generally at from 2 to 4% although catechins are contained in an amount as much as about 15%. As caffeine exhibits a central stimulant effect, it is used for the suppression of sleepiness. On the other hand, its excessive ingestion is considered to bring about adverse effects such as nervousity,

nausea and hyposomnia. Investigations have, therefore, been made for processes that selectively remove only caffeine from caffeine-containing compositions.

[0004]

Proposed are, for example, a process in which coffee is brought into contact with a caffeine adsorbent such as activated carbon under from 120 to 250 atm (Patent Document 3), and a process in which caffeine is selectively removed by bringing a caffeine-containing, aqueous solution into contact with activated clay or acid clay (Patent Document 4).

[0005]

However, the former relates to a supercritical extraction technology, requires an excessive process facility load, and lacks simplicity in its practice on industrial level. This process is accompanied by a further problem in that it does not only selectively remove caffeine alone but also changes the composition of catechins as an effective component. On the other hand, the latter process can selectively remove caffeine by simply using activated clay or acid clay, but is also accompanied by such a problem that the color deteriorates.

[0006]

[Patent Document 1] JP-A-60-156614

[Patent Document 2] JP-A-03-133928

[Patent Document 3] JP-A-53-18772

[Patent Document 4] JP-A-06-142405

[Disclosure of the Invention]

[Problem to Be Solved by the Invention]

[0007]

An object of the present invention is to provide a process for selectively removing caffeine from a green tea extract without significantly changing the composition of catechins and moreover, without deteriorating the color.

[Means for Solving the Problem]

[0008]

The present inventors have found that caffeine can be selectively removed from a green tea extract without significantly changing the composition of catechins, and moreover, without deteriorating the color by bringing, into contact with activated carbon and acid clay or activated clay, the green tea extract in a state that it is dispersed in a mixed solvent of water and an organic solvent at a specific ratio.

[0009]

The present invention provides a process for production of a purified green tea extract containing from 25 to 90 wt% of non-polymer catechins based on a dry weight of said extract, which comprises dispersing a green tea extract in a 91/9 to 97/3 by weight mixture of an organic solvent and water, and bringing the resulting dispersion into contact with activated carbon and acid clay or activated clay.

The present invention also provides a process for

selectively removing caffeine from a caffeine-containing catechin composition, which comprises dispersing a green tea extract in a 91/9 to 97/3 by weight mixture of ethanol and water and bringing the resulting dispersion into contact with activated carbon and acid clay or activated clay.

[Advantageous Effects of the Invention]

[0010]

According to the present invention, it is possible to selectively remove caffeine from a green tea extract without significantly changing the composition of catechins, and moreover, without changing the color. This process is particularly effective as a process for the production of a green tea extract which is high in the concentration of catechins but is low in the concentration of caffeine.

[Preferred Embodiments of the Invention]

[0011]

The green tea extract for use in the present invention contains one or more non-polymer catechins. The term "non-polymer catechins" as used herein is a generic term which collectively encompasses non-epicatechins such as catechin, gallocatechin, catechin gallate and gallocatechin gallate, and epicatechins such as epicatechin, epigallocatechin, epicatechin gallate and epigallocatechin gallate.

[0012]

As such green tea extracts containing non-polymer

catechins, those obtained by drying or concentrating extracts from green tea leaves can be mentioned.

[0013]

Tea leaves for use in the present invention include, more specifically tea leaves manufactured from tea leaves of the Genus *Camellia*, for example, *C. sinensis* and *C. assamica*, and the Yabukita variety or their hybrids. Such manufactured tea leaves include green teas such as sencha (middle-grade green tea), bancha (coarse green tea), gyokuro (shaded green tea), tencha (powdered tea) and kamairicha (roasted tea).

As an extraction method for obtaining a green tea extract from tea leaves, extraction can be conducted by a method such as stirring extraction. An organic acid or organic acid salt such as sodium ascorbate can be added beforehand to water upon extraction. It is also possible to make combined use of boiling deaeration or an extraction method which is conducted while flowing an inert gas such as nitrogen gas to eliminate dissolved oxygen, that is, under a so-called non-oxidizing atmosphere. The extract obtained as described above is dried to provide the green tea extract for use in the present invention. Forms of the green tea extract include liquid, slurry, semi-solid and solid forms. From the viewpoint of dispersibility in ethanol, a slurry, semi-solid or solid form is preferred.

[0014]

Instead of employing, as a green tea extract for use in

the present invention, a dried tea leaf extract, it is also possible to make use of a concentrate of a tea extract or to make combined use of a dried product of an extract from tea leaves and a concentrate of a tea extract.

The term "the concentrate of a tea extract" as used herein means a concentrate of an extract obtained from tea leaves with hot water or a water-soluble organic solvent, and includes, for example, those prepared by the processes disclosed in JP-A-59-219384, JP-A-04-20589, JP-A-05-260907, JP-A-05-306279 and the like.

As the green tea extract, it is possible to use specifically a commercially-available crude catechin preparation such as "Polyphenon" (product of Tokyo Food Techno Co., Ltd.), "TEAFURAN" (product of ITO EN, LTD.) or "SUNPHENON" (product of Taiyo Kagaku Co., Ltd.).

[0015]

As the green tea extract for use in the present invention, a concentrate of a green tea extract containing preferably from 25 to 90 wt%, more preferably from 25 to 70 wt%, still more preferably from 25 to 40 wt% of non-polymer catechins on a dry weight basis can be used preferably, because taste components other than the non-polymer catechins still remain.

[0016]

As the organic solvent for use in the present invention, ethanol, methanol, acetone, ethyl acetate or the like can be

mentioned. Among these, a hydrophilic organic solvent such as ethanol, methanol or acetone is preferred, with ethanol being more preferred in view of the use of the green tea extract in foods.

[0017]

In the present invention, the weight ratio of the organic solvent to water is adjusted to a range of from 91/9 to 97/3, preferably from 91/9 to 95/5, more preferably from 92/8 to 95/5. A proportion of the organic solvent greater than 97/3 results in a reduced extraction efficiency of catechins, while a proportion of the organic solvent smaller than 91/9 leads to a reduction in the degree of purification of the green tea extract.

[0018]

No particular limitation is imposed on the manner of dispersion of the green tea extract in the mixture of the organic solvent and water insofar as the weight ratio of the organic solvent to water upon final treatment of the caffeine-containing catechin composition is in the range of from 91/9 to 97/3.

[0019]

In the present invention, the treatment can be conducted by adding preferably from 10 to 40 weight parts, more preferably from 10 to 30 weight parts of the green tea extract to 100 weight parts of the mixture of the organic solvent and water, because the green tea extract can be efficiently treated.

[0020]



As to the time over which a required amount of water or the organic solvent is added, it is preferred to slowly add dropwise water or the organic solvent over a time of from approximately 10 to 30 minutes. To improve the efficiency of extraction of catechins, it is also preferred to effect the dropwise addition under stirring. It is more preferred to include an aging time of from 10 to 120 minutes or so after the completion of the dropwise addition of water.

These processing can be conducted at from 10 to 60°C, preferably from 10 to 50°C, more preferably from 10 to 40°C.

[0021]

No particular limitation is imposed on the activated carbon to be used in the present invention insofar as it is generally used on an industrial level. Usable examples include commercially-available products such as "ZN-50" (product of Hokuetsu Carbon Industry Co., Ltd.), "KURARAY COAL GLC", "KURARAY COAL PK-D" and "KURARAY COAL PW-D" (products of Kuraray Chemical K.K.), and "SHIROWASHI AW50", "SHIROWASHI A", "SHIROWASHI M" and "SHIROWASHI C" (products of Takeda Pharmaceutical Company Limited).

The pore volume of the activated carbon is preferably from 0.01 to 0.8 mL/g, more preferably from 0.1 to 0.7 mL/gm. The activated carbon having a specific surface area in a range of from 800 to 1,300 m<sup>2</sup>/g, particularly from 900 to 1,200 m<sup>2</sup>/g is preferred. It is to be noted that these physical values are

those determined by the nitrogen adsorption method.

[0022]

The activated carbon can be added preferably in a proportion of from 0.5 to 5 weight parts, particularly in a proportion of from 0.5 to 3 weight parts to 100 weight parts of the mixture of the organic solvent and water. The addition of activated carbon in an excessively small proportion leads to deteriorated decaffeination efficiency, while the addition of activated carbon in an unduly large proportion results in higher cake resistance in the filtration step. Proportions outside the above-described range are hence not preferred.

[0023]

Acid clay and activated clay for use in the present invention both contain, as general chemical components,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{MgO}$ , etc., and those having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of from 3 to 12, particularly from 4 to 9 are preferred. Also preferred are those of compositions which contain from 2 to 5 wt% of  $\text{Fe}_2\text{O}_3$ , from 0 to 1.5 wt% of  $\text{CaO}$  and from 1 to 7 wt% of  $\text{MgO}$ .

Activated clay is obtained by treating a naturally-occurring acid clay (montmorillonite clay) with a mineral acid such as sulfuric acid, and is a compound of a porous structure having large specific surface area and adsorbability. Further treatment of acid clay with an acid is known to change its specific surface area so that its decoloring ability is

improved and its physical properties are modified.

[0024]

The specific surface area of acid clay or activated clay is preferably from 50 to 350 m<sup>2</sup>/g although it varies depending on the degree of the acid treatment or the like, and its pH (5% suspension) is preferably from 2.5 to 8, particularly from 3.6 to 7. Usable examples of acid clay include commercially-available products such as "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.).

[0025]

Acid clay or activated clay can be added preferably in a proportion of from 2.5 to 25 weight parts, particularly in a proportion of from 2.5 to 15 weight parts to 100 weight parts of the mixture of the organic solvent and water. The addition of acid clay or activated clay in an unduly small proportion leads to a deterioration in decaffeination efficiency, while the addition of acid clay or activated clay in an excessively large proportion leads to an increase in the cake resistance in the filtration step. It is, therefore, not preferred to add acid clay or activated clay in a proportion outside the above-described range.

[0026]

The weight ratio of activated carbon to acid clay or activated clay can be from 1 to 10 of acid clay or activated clay to 1 of activated carbon, with the weight ratio of activated

carbon:acid clay or activated clay = 1:1 to 1:6 being preferred.

[0027]

The treatment of the dispersion of the green tea extract to contact it with activated carbon and acid clay or activated clay can be conducted by any method such as a batchwise treatment method or a continuous treatment method making use of a column. Adopted in general is a method in which powdery activated carbon or the like is added, the resulting mixture is stirred to selectively adsorb caffeine, and a filtering operation is then conducted to obtain a decaffeinated filtrate, or a method in which caffeine is selectively adsorbed by a continuous treatment using a column packed with granular activated carbon or the like.

After the contact with activated carbon and acid clay or activated clay, the dispersion of the green tea extract is subjected to distillation such as reduced-pressure distillation to remove the organic solvent from the system. The treated green tea extract can be either in a liquid form or in a solid form. To prepare it into a solid form, it may be formed into powder by a method such as freeze drying or spray drying.

[0028]

The purified green tea extract obtained by the present invention may preferably remain substantially unchanged in the composition of the non-polymer catechins contained therein compared with the composition before the treatment. The yield of the non-polymer catechins in the mixture of the organic solvent

and water before and after the treatment is preferably 70 wt% or more, more preferably 80 wt% or more.

[0029]

Further, it is preferred that in the non-polymer catechins in the purified green tea extract obtainable by the present invention, the ratio of gallocatechins, which consist of epigallocatechin gallate, gallocatechin gallate, epigallocatechin and gallocatechin, to non-gallocatechins, which consist of epicatechin gallate, catechin gallate, epicatechins and catechin, retains their composition in natural green tea leaves. Described specifically, from the standpoint that the purified product also retains their composition in natural green tea leaves, it is always preferred that the total amount of the above-described four gallocatechins is greater than that of the above-described four non-gallocatechins.

[0030]

Furthermore, the content of the gallates consisting of catechin gallate, epicatechin gallate, gallocatechin gallate and epigallocatechin gallate in the purified green tea extract obtained by the present invention is preferably 45 wt% or higher of the whole non-polymer catechins from the standpoint of the effectiveness of physiological effects of the non-polymer catechins.

[0031]

The concentration of caffeine in the purified green tea

extract obtained by the treatment according to the present invention is preferably from 25 to 200, more preferably from 30 to 150, still more preferably from 30 to 100 relative to the non-polymer catechins, that is, in terms of the non-polymer catechins/caffeine (by weight).

[0032]

The purified green tea extract obtained by the present invention may contain preferably from 25 to 90 wt%, more preferably from 25 to 70 wt%, still more preferably from 40 to 70 wt% of the non-polymer catechins. In the solid content of the purified green tea extract, the non-polymer catechins may amount preferably to from 40 to 90 wt%, more preferably to from 50 to 90 wt%, still more preferably to from 60 to 90 wt%.

[0033]

The resulting purified green tea extract, despite its low caffeine concentration, still contains non-polymer catechins at a high concentration, has a good color, and further, is substantially free of the flavor of green tea. Therefore, packaged beverages in which the purified green tea extract is incorporated are useful particularly as non-tea beverages such as sports drinks and isotonic drinks.

[0034]

The packaged beverage according to the present invention contains non-polymer catechins (A) dissolved in water in an amount of preferably from 0.03 to 1.0 wt%, more preferably from

0.04 to 0.5 wt%, yet more preferably from 0.06 to 0.4 wt%, still more preferably from 0.07 to 0.4 wt%, even more preferably from 0.08 to 0.3 wt%, still even more preferably from 0.09 to 0.3 wt%, yet still even more preferably from 0.1 to 0.3 wt%. Insofar as the content of non-polymer catechins falls within the above-described range, a great deal of non-polymer catechins can be taken with ease, and from the standpoint of the color tone of the beverage shortly after its preparation, this content range is also preferred. The concentration of the non-polymer catechins can be adjusted by the amount of the green tea extract to be incorporated.

Further, the daily intake required for an adult to exhibit the effects of the promotion of accumulated fat burning, the promotion of dietary fat burning and the promotion of  $\beta$ -oxidation gene expression in the liver is preferably 300 mg or more, more preferably 450 mg or more, still more preferably 500 mg or more in terms of non-polymer catechins. Specifically, it has been confirmed that an anti-puffiness effect and/or visceral fat reducing effect can be brought about by ingesting a beverage which contains 483 mg, 555 mg or 900 mg of non-polymer catechins per package (JP-A-2002-326932).

Therefore, the daily intake of the packaged beverage according to the present invention for an adult can also be preferably 300 mg or more, more preferably 450 mg or more, still more preferably 500 mg or more in terms of non-polymer catechins.

From the standpoint of assuring the required daily intake amount, the non-polymer catechins may be contained in an amount of preferably 300 mg or more, more preferably 450 mg or more, still more preferably 500 mg or more per package of the packaged beverage according to the present invention.

[0035]

The weight ratio of the non-polymer catechins to caffeine contained in the packaged beverage according to the present invention is preferably from 25 to 200, more preferably from 30 to 150, still more preferably from 30 to 100.

[0036]

The packaged beverage according to the present invention may also contain sodium ions and/or potassium ions. Beverages of the present invention containing such ions are useful in the form of drinks such as sports drinks and isotonic drinks. The term "sports drink" is generally defined to mean a drink that can promptly replenish water and minerals lost in the form of sweat during physical exercise.

[0037]

Sodium and potassium can be mentioned as primary physiological electrolytes. These ion ingredients can be incorporated by adding in their corresponding water-soluble ingredients or inorganic salts. They are also found in fruit juices and tea extracts. The amount of an electrolyte or ion ingredient in the beverage according to the present invention



is its content in the final packaged beverage ready for drinking. The concentration of an electrolyte is expressed in terms of "ion concentration". In the beverage according to the present invention, a potassium ion ingredient can be added in the form of a salt such as potassium chloride, potassium carbonate, potassium sulfate, potassium acetate, potassium hydrogencarbonate, potassium citrate, potassium phosphate, potassium hydrogenphosphate, potassium tartrate, potassium sorbate or a mixture thereof or as a component of an added fruit juice or tea. In the packaged beverage according to the present invention, potassium ions can be contained in an amount of preferably from 0.001 to 0.2 wt%, more preferably from 0.002 to 0.15 wt%, even more preferably from 0.003 to 0.12 wt%. Similarly, a sodium ion ingredient can also be added as a readily-available sodium salt such as sodium chloride, sodium carbonate, sodium hydrogencarbonate, sodium citrate, sodium phosphate, sodium hydrogenphosphate, sodium tartrate, sodium benzoate or a mixture thereof or as a component of an added fruit juice or tea. A lower sodium ion concentration is desired from the standpoint of facilitating the absorption of water owing to osmotic pressure. Preferably, however, the sodium ion concentration should be of such a level as to avoid suction of water into the intestine from the body by osmotic pressure. The sodium ion concentration required to achieve such a level can preferably be lower than the sodium ion concentration in the

plasma. In the packaged beverage according to the present invention, sodium ions can be contained at preferably from 0.001 to 0.5 wt%, more preferably from 0.002 to 0.4 wt%, still more preferably from 0.003 to 0.2 wt%. In addition to potassium and sodium ions, from 0.001 to 0.5 wt%, preferably from 0.002 to 0.4 wt%, still more preferably from 0.003 to 0.3 wt% of chloride ions can also be incorporated in the packaged beverage according to the present invention. A chloride ion ingredient can be added in the form of a salt such as sodium chloride or potassium chloride. Further, trace ions such as calcium, magnesium, zinc and/or iron can also be added. These ions can also be added in the form of salt or salts. The total amount of ions existing in the beverage includes not only an amount of ions added but also an amount of ions naturally existing in the beverage. When sodium chloride is added, for example, the amounts of sodium ions and chloride ions in the added sodium chloride are included in the total amount of respective ions accordingly.

Depending upon the situation of drinking, an excessively low concentration of sodium ions and potassium ions may fail to provide a fulfilled feeling in taste and to achieve an effective replenishment of minerals, and therefore, may not be preferred. An unduly high concentration of sodium ions and potassium ions, on the other hand, leads to strong tastes of the salts themselves and is not preferred for long-term drinking.

[0038]

In the packaged beverage according to the present invention, a sweetener may also be used to improve the taste. As the sweetener, an artificial sweetener, carbohydrate or glycerol (for example, glycerin) can be used. The content of such a sweetener in the packaged beverage according to the present invention is preferably from 0.0001 to 20 wt%, more preferably from 0.001 to 15 wt%, even more preferably from 0.001 to 10 wt%. A content lower than the above-described lower limit cannot impart much sweetness and cannot achieve a balance between sourness and saltiness, while a content higher than the above-described upper limit leads to excessive sweetness and therefore, to the strong feeling of being caught in the throat and a reduction in the feeling as the beverage passes down the throat.

As the sweeteners usable in the packaged beverage according to the present invention, use of an artificial sweetener is preferred. Example of the artificial sweetener usable in the present invention include high-sweetness sweeteners such as saccharin, saccharin sodium, aspartame, acesulfame-K, sucralose and neotame; and sugar alcohols such as sorbitol, erythritol and xylitol. As commercial products, "SLIM-UP SUGAR" composed of aspartame, "LAKANTO-S" containing erythritol, and "PALSWEET" composed of erythritol and aspartame can be used.

[0039]

When the aimed packaged beverage is also intended to

replenish energy, it is preferred to use a carbohydrate sweetener.

As the carbohydrate sweeteners usable in the present invention, soluble carbohydrates can be employed. A soluble carbohydrate serves not only as a sweetener but also as an energy source. Upon choosing a carbohydrate for use in the beverage according to the present invention, it is necessary to take a sufficient gastric excretion rate and intestinal absorption rate into consideration.

[0040]

The carbohydrate can be a mixture of glucose and fructose, or a carbohydrate hydrolyzable into glucose and fructose or capable of forming glucose and fructose in the digestive tract. The term "carbohydrate" as used herein includes monosaccharides, oligosaccharides, conjugated polysaccharides, and mixtures thereof.

[0041]

Monosaccharides usable in the present invention include tetroses, pentoses, hexoses and ketohexoses. Examples of the hexoses are aldohexoses such as glucose known as grape sugar. The content of glucose in the packaged beverage according to the present invention can be preferably from 0.0001 to 20 wt%, more preferably from 0.001 to 15 wt%, still more preferably from 0.001 to 10 wt%. Fructose known as fruit sugar is a ketohexose. The content of fructose in the packaged beverage according to

the present invention can be preferably from 0.0001 to 20 wt%, more preferably from 0.001 to 15 wt%, particularly preferably from 0.001 to 10 wt%.

Preferred in the beverage according to the present invention is a single artificial sweetener system or a combination of an artificial sweetener and a glucose compound or a combination of an artificial sweetener and a fructose compound.

[0042]

As a carbohydrate sweetener for use in the present invention, a soluble carbohydrate can be employed. As an oligosaccharide, a carbohydrate which produces these two kinds of monosaccharides in vivo (specifically, sucrose, maltodextrin, corn syrup, and fructose-rich corn syrup) can be mentioned. It is a disaccharide that is an important type of the oligosaccharide. An illustrative disaccharide is sucrose known as cane sugar or beet sugar. The content of sucrose in the packaged beverage according to the present invention can be preferably from 0.001 to 20 wt%, more preferably from 0.001 to 15 wt%, particularly preferably from 0.001 to 10 wt%.

[0043]

The pH of the packaged beverage according to the present invention can be preferably from 2 to 6, more preferably from 2 to 5, still more preferably from 3 to 4.5 from the standpoint of the stability of catechins. An excessively low pH provides

the beverage with a stronger sour taste and pungent smell. An unduly high pH, on the other hand, fails to achieve a harmony in flavor and leads to reduction in taste. Such an excessively low pH or unduly high pH is, therefore, not preferred.

[0044]

Addition of a bitterness suppressor to the packaged beverage according to the present invention facilitates its drinking, and therefore, is preferred. Although no particular limitation is imposed on the bitterness suppressor to be used, a cyclodextrin is preferred. As the cyclodextrin, an  $\alpha$ -,  $\beta$ - or  $\gamma$ -cyclodextrin or a branched  $\alpha$ -,  $\beta$ - or  $\gamma$ -cyclodextrin can be used. In the beverage, a cyclodextrin may be contained preferably in an amount of from 0.005 to 0.5 wt%, more preferably from 0.01 to 0.3 wt%. To the packaged beverage according to the present invention, it is possible to add, either singly or in combination, additives such as antioxidants, flavorings, various esters, organic acids, organic acid salts, inorganic acids, inorganic acid salts, inorganic salts, dyes, emulsifiers, preservatives, seasoning agents, sweeteners, sour seasonings, gums, emulsifiers, oils, vitamins, amino acids, fruit extracts, vegetable extracts, flower honey extracts, pH regulators and quality stabilizers.

[0045]

As in general beverages, a package used for the packaged beverage according to the present invention can be provided in

a conventional form such as a molded package made essentially of polyethylene terephthalate (a so-called PET bottle), a metal can, a paper container combined with metal foils or plastic films, a bottle or the like. The term "packaged beverage" as used herein means a beverage that can be taken without dilution.

[0046]

The packaged beverage according to the present invention can be produced, for example, by filling the beverage in a package such as a metal can and, when heat sterilization is feasible, conducting heat sterilization under sterilization conditions as prescribed in the Food Sanitation Act. For packages which cannot be subjected to retort sterilization such as PET bottles or paper packages, adopted is a process in which the beverage is sterilized beforehand at a high temperature for a short time under similar sterilization conditions as those described above, for example, by a plate-type heat exchanger or the like, is cooled to a predetermined temperature, and is then filled in a package. Under aseptic conditions, additional ingredients may be added to and filled in a beverage-filled package. It is also possible to conduct such an operation that subsequent to heat sterilization under acidic conditions, the pH of the beverage is brought back to neutral under aseptic conditions or that subsequent to heat sterilization under neutral conditions, the pH of the beverage is brought back to acidic under aseptic conditions.

[Examples]

[0047]

Measurement of catechins

A high-performance liquid chromatograph (model: "SCL-10AVP") manufactured by Shimadzu Corporation was fitted with a liquid chromatograph column packed with octadecyl-introduced, "L-Column, TM ODS" (4.6 mm in diameter × 250 mm; product of Chemicals Evaluation and Research Institute, Japan). A purified green tea extract was diluted with distilled water, filtered through a filter (0.8 μm), and then subjected to chromatography at a column temperature of 35°C by the gradient elution method making use of a solution A and a solution B. The solution A, mobile phase, was a solution containing 0.1 mol/L of acetic acid in distilled water, while the solution B was a solution containing 0.1 mol/L of acetic acid in acetonitrile. The measurement was conducted under the conditions of 20 μL sample injection volume and 280 nm UV detector wavelength.

[0048]

Measurement of caffeine

(Analyzer)

A HPLC system (manufactured by Hitachi, Ltd.) was used. Plotter: "D-2500", Detector: "L-4200", Pump: "L-7100", Autosampler: "L-7200", Column: "Intersil ODS-2" (2.1 mm in inner diameter × 250 mm in length).

(Analytical conditions)



Sample injection volume: 10  $\mu$ L

Flow rate: 1.0 mL/min

Detection wavelength of UV spectrophotometer: 280 nm

Eluent A: A 0.1 M solution of acetic acid in water

Eluent B: A 0.1 M solution of acetic acid in acetonitrile

Concentration gradient conditions (vol. %)

Time	Eluent A	Eluent B
0 min	97%	3%
5 min	97%	3%
37 min	80%	20%
43 min	80%	20%
43.5 min	0%	100%
48.5 min	0%	100%
49 min	97%	3%
62 min	97%	3%

(Retention time of caffeine)

Caffeine: 27.2 min

From each area % determined here, the corresponding wt% was determined based on the standard substance.

[0049]

#### Assessment of color

(Analyzer)

"UV MINI1240" (manufactured by Shimadzu Corporation) analyzer was used.

An absorbance value at 450 nm was measured by a

spectrophotometer. In the measurement, each purified green tea extract was diluted with ion-exchanged water such that the concentration of catechins was lowered to 100 mg%. Using the sample, the absorbance was measured, and was employed as an index of color.

#### Visual assessment of stability

Each purified green tea extract was diluted with ion-exchanged water such that the concentration of catechins was lowered to 100 mg%. An assessment sample filled in a 50-mL vial was observed for the state of its contents on an illuminator, and the state of the contents was visually ranked.

[0050]

[Table 1]

	Example	Example	Comp. Ex.	Comp. Ex.
	1	2	1	2
Solid green tea extract (g) ("POLYPHENON HG", product of Tokyo Food Techno Co., Ltd.)	200	100	100	100
Ethanol (g)	760	830	0	0
Water (g)	40	70	900	900
Activated carbon (g) ("KURARAY COAL GLC" (product of Kuraray Chemical K.K.))	20	25	0	20
Acid clay (g) "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.)	100	30	100	100
Organic solvent/water (weight ratio)	95/5	92/8	0/100	0/100
Non-polymer catechins after treatment (wt%) <sup>1)</sup>				
GC	6.26	6.25	6.75	7.42
EGC	29.71	29.72	31.75	34.72
C	2.00	1.92	2.22	2.02
EC	9.92	10.01	9.64	8.24
EGCg	37.65	37.65	35.93	35.86
GCg	1.26	1.23	1.39	1.47
ECg	12.14	12.19	11.34	9.61
C	1.08	1.02	0.94	0.66
Non-polymer catechins/caffeine after treatment (weight ratio)	33.0	59.5	23.6	42.6
Gallates percentage of non-polymer catechins after treatment (wt%)	51.0	51.2	49.6	47.6
Gallocatechins percentage of non-polymer catechins after treatment (wt%)	74.9	73.5	75.8	79.5
Concentration of non-polymer catechins in solids after treatment (wt%)	66	64	34	34
Absorbance (-)	0.038	0.031	0.535	0.270
Assessment of purified products	Caffeine content was lowered, color was good, and stability was visually good.	Caffeine content was lowered, color was good, and stability was visually good.	Color deteriorated, and precipitation occurred.	Color deteriorated, and precipitation occurred.

1) Composition of non-polymer catechins in the "POLYPHENON HG" preparation:

GC (gallocatechin) 6.39%, EGC (epigallocatechin) 29.42%, C (catechin) 2.16%, EC (epicatechin) 10.3%, EGCg (epigallocatechin gallate) 37.13%, GCg (gallocatechin gallate) 1.93%, ECG (epicatechin gallate) 11.89%, Cg (catechin gallate) 0.79%, gallates percentage 51.73%, gallocatechins percentage 74.88%

[0051]

As shown in Table 1, the combined use of activated carbon and acid clay while employing an aqueous solution, which contains an organic solvent at a very high concentration, enables production of a purified green tea extract having lowered caffeine content and good color and stability without changing the gallates percentage and gallocatechins percentage of its non-polymer catechins.

[0052]

(Example 1)

\*1: Green Tea Extract A

A green tea extract ("POLYPHENON HG", product of Tokyo Food Techno Co., Ltd.; 200 g) was dispersed in a 95% aqueous ethanol solution (800 g) at room temperature under stirring at 250 rpm. After addition of acid clay "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.; 100g), stirring was continued for about 10 minutes. Subsequently, filtration was conducted through No.2 filter paper. Activated carbon (20 g) was then added, followed by filtration through No.2 filter paper again. Re-filtration was then conducted through a 0.2- $\mu$ m membrane filter. Finally, ion-exchanged water (200 g) was added to the filtrate, ethanol was distilled away at 40°C and 0.0272 kg/cm<sup>2</sup>, and the concentration of catechins was adjusted with ion-exchanged water to obtain a product.

Content of non-polymer catechins

after the treatment: 22 wt%.

Non-polymer catechins/caffeine weight

ratio after the treatment: 33.0

Gallates percentage after the treatment: 51 wt%.

(Example 2)

\*2: Green Tea Extract B

A green tea extract ("POLYPHENON HG", product of Tokyo Food Techno Co., Ltd.; 100 g) was dispersed in a 70% aqueous ethanol solution (100 g) at room temperature under stirring at 250 rpm. After addition of activated carbon "KURARAY COAL GLC" (product of Kuraray Chemical K.K.; 25 g) and acid clay "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.; 30g), stirring was continued for about 10 minutes. After a 95% aqueous ethanol solution (800 g) was added dropwise over 30 minutes, stirring was continued for 30 minutes at room temperature. Subsequently, filtration was conducted again through No.2 filter paper. Re-filtration was then conducted through a 0.2- $\mu$ m membrane filter. Finally, ion-exchanged water (200 g) was added to the filtrate, ethanol was distilled away at 40°C and 0.0272 kg/cm<sup>2</sup>, and the water content was adjusted to obtain a desired product.

Content of non-polymer catechins

after the treatment: 22 wt%.

Non-polymer catechins/caffeine weight

ratio after the treatment: 59.5

Gallates percentage after the treatment: 51.2 wt%.

(Comparative Example 1)

"POLYPHENONHG" (product of Tokyo Food Techno Co., Ltd.; 100 g) was dispersed as a green tea extract in water (900 g) at room temperature under stirring at 250 rpm. After the addition of acid clay "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.; 100g), stirring was continued for about 20 minutes. Subsequently, stirring was continued for about 30 minutes at room temperature. After filtration was conducted through No.2 filter paper, re-filtration was conducted through a 0.2- $\mu$ m membrane filter. Finally, water was gradually evaporated by a drying apparatus until the concentration of non-polymer catechins became equivalent to that in Example 1 to obtain a product.

Content of non-polymer catechins

after the treatment: 22 wt%.

Non-polymer catechins/caffeine weight

ratio after the treatment: 23.6

Gallates percentage after the treatment: 49.6 wt%.

(Comparative Example 2)

"POLYPHENONHG" (product of Tokyo Food Techno Co., Ltd.; 100 g) was dispersed as a green tea extract in water (900 g) at room temperature under stirring at 250 rpm. After the addition of activated carbon "KURARAY COAL GLC" (product of

Kuraray Chemical K.K.; 20 g) and acid clay "MIZUKA ACE #600" (product of Mizusawa Chemical Industries, Ltd.; 100g), stirring was continued for about 20 minutes. Stirring was then continued for about 30 minutes at room temperature. After filtration was conducted through No.2 filter paper, re-filtration was conducted through a 0.2- $\mu$ m membrane filter. Finally, water was gradually evaporated by a drying apparatus until the concentration of non-polymer catechins became equivalent to that in Example 1 to obtain a product.

Content of non-polymer catechins

after the treatment: 22 wt%.

Non-polymer catechins/caffeine weight

ratio after the treatment: 42.6

Gallates percentage after the treatment: 47.6 wt%.

[0053]

<Production Procedure of Packaged Beverages of Examples 1-2  
& Comparative Example 1>

Formulated solutions were prepared by adding the respective green tea extracts shown in Table 1, and then bringing the total amounts to 100 with ion-exchanged water, respectively. The formulated solutions were subjected to a sterilization step according to the Food Sanitation Act and hot-pack filling to obtain packaged beverages.

[0054]

[Table 2]

Formulations	Example 1	Example 2	Comparative Example 1
Green tea extract A	1.00	-	-
Green tea extract B	-	1.00	-
Green tea extract C	-	-	0.65
Antioxidant	0.030	0.030	0.030
Sour seasoning	0.300	0.300	0.300
Sweetener	5.000	5.000	5.000
Fruit juice	0.050	0.050	0.050
Ion-exchanged water	Balance	Balance	Balance
Total amount	100	100	100
pH of beverage	3.5	3.5	3.6
Non-polymer catechins (wt%)	0.22	0.22	0.22
Non-polymer catechins/caffeine ratio	33	59.5	6.1
Mouth refreshment shortly after drinking	4	5	1



[0055]

The green tea extract C added in Table 2 had the following composition:

\*3: Green Tea Extract C

Caffeine-containing catechin composition ("POLYPHENON HG", product of Tokyo Food Techno Co., Ltd).

Content of non-polymer catechins 33.70 wt%, caffeine content 5.5 wt%, non-polymer catechins/caffeine: 6.1, gallates percentage 51 wt%.

[0056]

Five trained male assessors were used. Those assessors ingested 350 mL of each beverage, and assessed the beverages by giving ranking scores to their mouth refreshment shortly after drinking in accordance with the following standards.

[0057]

- 5: Very good mouth refreshment
- 4: Good mouth refreshment
- 3: Slightly good mouth refreshment
- 2: Slightly poor mouth refreshment
- 1: Poor mouth refreshment

[0058]

As evident from the results of Table 2, a packaged beverage with outstanding mouth refreshment shortly after its drinking can be obtained by using an invention product in which, while maintaining the composition of catechins, caffeine has

been selectively removed by treating a green tea extract in accordance with the present invention.

[Document Name] ABSTRACT

[Abstract]

[Problem] To provide a process for selectively removing caffeine from a green tea extract without significantly changing the composition of catechins, and moreover, without changing the color.

[Solution] A process for production of a purified green tea extract containing from 25 to 90 wt% of non-polymer catechins based on a dry weight of said extract, which comprises dispersing a green tea extract in a 91/9 to 97/3 by weight mixture of an organic solvent and water, and bringing the resulting dispersion into contact with activated carbon and acid clay or activated clay.

[Chosen Drawing] None